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USSR TRENDS IN WORK ON HYDROCARBONS AND THE COMBUSTION OF HYDROCARBONS

[Comment: This report reviews recent USSR research on hydrocarbons and technical developments based on research of this nature as applied to diesel fuels and related fuels, such as tractor kerosene and turbojet fuel. An effort to broaden the crude-material basis for the production of fuels of this range is apparent in the work reviewed. Some of the research summarized emphasizes low pour points (a critical characteristic of turbojet fuels) rather than high cetane numbers, which are more important for diesel fuel applications. The discussion of the prospects of producing special fuels synthetically on an industrial scale is of particular interest. From this standpoint, the hydropolymerization of butadiene apparently offers the best possibilities.

Numbers in parentheses refer to appended sources.]

Purpose of Current Research

USSR authors state that increasing attention must be paid in that country to work on hydrocarbon fuels of the composition C_{12} - C_{25} and have published results of recent work done by them along these lines.(1, 4)

In his book on motor fuels, A. D. Petrov summarizes the results of a systematic investigation of hydrocarbons carried out at the Institute of Organic Chemistry, Academy of Sciences USSR, and the Chair of Organic Chemistry, Gor'kiy State University, with the view of determining the composition of hydrocarbons which have the most desirable characteristics as diesel fuels. He states that in research of this type hydrocarbons ought to be found which have pour points of minus 50° and lower, and cetene numbers lying within the medium range of 40-50, because the designs of diesel engines

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which are available at present do not warrant the use of fuels having cetene values higher than that. He adds that hydrocarbons having a pour point within the desired low range, but cetene numbers which are much lower than 40-50, can be used in air reaction engines of the gas turbine type [e.g., turbojets]. As far as fulfillment of requirements for diesel fuels is concerned, Petrov states that the isoparaffins are the most satisfactory class of hydrocarbons in that respect, because they not only have low pour points, but also exhibit satisfactory temperatures of spontaneous ignition, i.e., ignite at low compression ratios. Petrov further says that isoparaffins have a low viscosity, which makes possible inclusion of isoparaffinic hydrocarbons of higher molecular weights (corresponding to an upper limit of 24 carbon atoms or more in the molecule) as compared with straight chain paraffins or aromatics, into the composition of diesel fuels and broadens in a similar manner the fraction that can be used as gas turbine fuel, i.e., in a type of application which requires still lower viscosities. He suggests that the relatively low tendency of isoparaffins to oxidize, which is the only drawback of this class of hydrocarbons, may be improved by adding ethers, peroxides, or nitrocompounds.

He further states that naphthenic hydrocarbons, which have the same undesirable viscosity characteristics as aromatic compounds, are characterized by higher cetene numbers and lower pour points than the corresponding aromatic hydrocarbons, so that the latter can be improved by subjecting them to low-temperature hydrogenation and thus converting them into naphthenes. He also recommends that industrial synthesis of diesel fuel additives which lower the pour point be carried out by alkylating aromatic hydrocarbons and thereby introducing a long-branched chain into the benzene nucleus or a short chain into naphthalene to synthesize monoalkyldecahydronaphthalenes. According to Petrov, another possibility of synthesizing diesel fuel pour point depressants is polymerization of normal olefins (e.g., n-butene) with the aid of catalysts which do not produce isomerization, or polymerization of such olefins by means of electric discharges.(1)

Work on the polymerization of unsaturated hydrocarbons by means of electric discharges has been conducted by USSR workers. A summary of research on this subject is contained in a monograph written by D. N. Andreyev.(5)

Industrial Synthesis of Special Fuels

In discussing the correlation between chemical constitution and low pour points in potential components of diesel fuels within the C_{12} - C_{24} range, A. D. Petrov outlines the properties of (I) hydrocarbons possessing a structure of the 2,2,4,6,6-pentamethylheptane (hydrogenated triisobutylene) type; (II) hydrocarbons having a somewhat different (T-shaped) structure of the trialkylmethane type (e.g., tributylmethane); (III) hydrocarbons of the tetraalkylethane type and (IV) hydrocarbons which have a structure of the "comb" type (e.g. triethyltetradecane). After pointing out that hydrocarbons of the type II, which have only one tertiary carbon atom [and lack the quaternary carbon atoms that are characteristic for type I], exhibit a maximum oxidability which is close to that of normal paraffins and results in high cetene numbers, while at the same time the pour points are low by reason by the asymmetric structure of the hydrocarbons, Petrov states that convenient methods for the industrial synthesis of hydrocarbons of this type are not available as yet. He goes on to say that application of the Grignard synthesis for the production of hydrocarbons of this class on an industrial scale is not out of the question.

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However, he points out, that notwithstanding the ready availability of the starting materials for this type of synthesis (oleic and enanthic acids and primary alkyl chlorides such as hexyl chloride and nonyl chloride), which are obtained by the distillation and hydrogenation of natural and synthetic fatty acids, or derived from distillation and hydrogenation products obtained in this manner, the industrial Grignard synthesis of components of diesel fuels will be prohibitive from the standpoint of cost, at least in the near future, although cheap magnesium may be available. According to Petrov, application of the Grignard synthesis (in combination with a Wuertz synthesis) will likewise not pay in the industrial production of hydrocarbons of type III, which furthermore have low knock stability as compared with hydrocarbons of type II. Petrov states that a two-step process, consisting of a Grignard synthesis followed by a Wuertz synthesis, is feasible, as far as cost considerations are concerned, only for the industrial production of special lubricants.(1)

Elsewhere in the book, Petrov states that it might be possible to produce industrially by means of the Grignard synthesis both hydrocarbon diesel fuel components and hydrocarbon lubricating oils at installations similar to those used for the production of silicones, and under the same conditions.(2)

After discussing hydrocarbons of type III, Petrov says that the most promising hydrocarbons, from the standpoint of the industrial synthesis of diesel oil components, are those of type IV, because they can be prepared in a simple manner by polymerizing butadiene at low temperatures in the presence of butyl-lithium (i.e. by applying a variation of the Ziegler synthesis), or by subjecting butadiene to hydropolymerization. He adds that while the anti-knock properties of this class of hydrocarbons have not been investigated as yet, their pour points and cetene numbers proved to be very satisfactory, the latter amounting to about 80 for hydrocarbons having pour points below minus 60°C. According to Petrov, the pour points and cetene number of synthetic hydrocarbons of this type are superior by a factor of two to those shown by products of the selective purification of petroleum gas oils after paraflow had been added to the refined products of this purification, i.e., fractions from which aromatics and paraffins having high pour points had been removed.(1)

Low Pour Points Emphasized

In Petrov's discussion of the chemistry of diesel fuels and of other fuels which are close to them in fractional composition, the problem of attaining low pour points is emphasized. N. I. Chernozhukov's handbook of petroleum technology states that the pour point of the fuel is of great importance in the operation of high speed engines: according to USSR technical specifications, summer diesel fuel for automobile and tractor engines should have a pour point no higher than minus 10°C, winter diesel fuel for the same types of engines a pour point no higher than minus 35°C, and aviation diesel fuel a pour point no higher than minus 60°C. This book furthermore states that low pour points are of particular importance in fuel that is to be used in turbocompressor air reaction engines [turbojets], because low starting temperatures may be encountered in the winter and the engines must operate at low temperatures of the surrounding air. The desirable characteristics of turbojet fuels are outlined in some detail in this book (6) on the basis of data contained in Neftyanoye Tovarovedeniye (Petroleum Products) by B. V. Losikov and I. P. Lukashevich. According to Losikov and Lukashevich, turbojet fuel must remain perfectly fluid at minus 50°C.(7) Chernozhukov repeats this statement and amplifies it by citing specifications for turbojet fuel from the US periodical Petroleum Processing, Oct 1946, p. 45. These specifications stipulate that the pour point of turbojet fuel must not exceed minus 60°C.(6)

Losikov and Lukashevich state that the petroleum fraction corresponding to kerosene [C_{16} - C_{20}] forms the best turbojet fuel, and add that although a fraction consisting primarily of paraffin hydrocarbons is best for this type of application, fuels having a high content of aromatic hydrocarbons can be used if the combustion is conducted in such a manner that the chief drawbacks of aromatics are eliminated. These drawbacks are a high ignition temperature and formation of carbon particles which may cause erosion of the turbine blades.(7) According to Petrov, aromatic hydrocarbons are very undesirable constituents of air reaction engine fuels.(1)

Petrov on Current Developments

In the introduction to his book on motor fuels, Petrov outlines the interrelationships between diesel fuels, gas turbine fuels, and related types of fuels in the following general terms, giving some attention to the effects exerted by aromatic constituents of some of these fuels:

The application of methods of complete conversion to gasoline (ordinary cracking and hydrogenation cracking) has resulted in a better utilization of crude petroleum and synthetic liquid crude materials. Extensive use of diesel engines and lately of air reaction engines (gas turbines, turbojets) and rocket engines has contributed to a still more complete utilization of these crude materials. The solar oils and fuel oils ["motor petroleum"] used as diesel fuel at the same time represent crude material for cracking. Diesel fuel and particularly diesel fuel which has a low autoignition point must satisfy certain specific requirements in regard to its quality. The degree of knocking of diesel engines depends on the inflammability of the fuel used in them. The use of a fuel that ignites readily results in a smooth operation of the diesel engine. It has been established that reduction of the induction period of ignition results in a smooth operation of the diesel engine and elimination of knocking.

All substances which reduce the induction period of ignition (e.g., amyl nitrite, benzaldehyde, and acetaldehyde) function as effective antiknock substances when added to a diesel fuel. On the other hand, antiknock agents for gasoline (e.g., tetraethyl lead and benzene) are not acceptable as additives to diesel fuel, because they function as knock promoters when added to this fuel.

Fuels consisting mainly of normal paraffins produce the strongest knocking in carburetor motors. On the other hand, paraffin fuel can be used most effectively in diesel engines. For instance, diesel fuels derived from Surakhan and Grozny crudes insure smooth operation of the engines. This is due to the fact that paraffinic fuels have a relatively low autoignition temperature.

When different types of fuel are combined, a fuel which has the desired cetene number can be obtained. For instance, a diesel fuel that was used rather extensively in Germany consisted of a coal-tar fraction having a cetene number lower than 10 and a synthin fraction having a cetene number of 100 (synthin consists of synthetic, mostly normal hydrocarbons of the paraffin and olefin series which have been formed by reacting carbon monoxide with hydrogen over a cobalt catalyst).

As compared with carburetor engines, diesel engines are not used extensively, particularly in Western Europe and the US. However, one must consider in this connection the expanding manufacture of diesel engines and the fact that the motor fuels which are available in the USSR are to some extent different from those which are available abroad.

50X1-HUM

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The socialist economy of the USSR has created extensive possibilities for the use of tractors, motorbuses, and motor trucks. The development of freight transportation by motor truck has stimulated the manufacture of diesel engines. This in turn has resulted in extensive attention to work on fuels possessing suitable characteristics for light diesel engines.

The kerosene fraction (200-300°C) and the ligroin-kerosene fraction (65-300°C) are required not only for diesel engines, but also for the jet and rocket engines which were developed at the end of World War II. While the paraffin hydrocarbons of gasoline synthesized from water gas were used primarily in the first type of engine [jet engines], engines of the second type [rocket engines] were operated by using either "arobin" (an aromatic gasoline containing more than 40% of aromatic hydrocarbons), or the xylene fraction of coal tar, or aromatized gasoline obtained by the destructive hydrogenation of coal. The rocket engine fuel component of any of the three types mentioned was combined with an amine, catechol, or some other initiator of ignition. Ignition was produced by mixing the fuel with nitric acid. According to some directions, mixtures of alcohols (methyl alcohol mixed with ethyl alcohol) and liquid oxygen or hydrogen peroxide were used.

One must also remark that while prior to World War II fuel rich in isoparaffins was considered best for aviation engines using the Otto cycle, frequent use of engines under conditions of exceptional strain during the war has contributed to the development of engines equipped with superchargers and in consequence thereof to an increased demand for fuels rich in aromatic hydrocarbons.

Petrov concludes this discussion by saying that the increased demand for aromatic hydrocarbons (and other aromatic compounds such as amines and phenols) for modern motor fuels and propellants combined with the demand for aromatic hydrocarbons to be used in the explosives and synthetic rubber industries has not only resulted in a retention of the coal liquefaction industry by countries which lack petroleum resources, such as Germany and Italy, but has also stimulated the development of this industry in countries where plentiful supplies of crude petroleum exist.(3)

Specifications for Diesel Fuel

In discussing diesel fuels, Losikov and Lukashevich state that up to now this type of fuel consisted chiefly of straight-run distillates. They point out, however, that gas oil fractions comprise 40-48% of the products resulting from catalytic cracking and that gas oil derived from cracking will form an increasing proportion of the total amount of diesel oil as the application of catalytic cracking expands. They outline the characteristics of the five standard grades of diesel fuel which are being supplied presently by the USSR petroleum industry. This information is given in the attached table.(7) In outlining the properties of diesel fuels, Chernozhukov says that light diesel fuels must have a cetane number of at least 50, and that light diesel fuels of the [straight-run] distillate type having a strictly limited boiling temperature range are now used predominantly in high-speed diesels.(6)

Broadening of Raw Material Basis

A recent article published in a USSR periodical states that the increased production and use of high-speed diesel engines raises the question of extending the range of raw materials used for the production of diesel fuels. According to the authors of the article, it is necessary to broaden the rather narrow kerosene-gas oil fraction from which fuel for high-speed diesels was derived hitherto and which is not available in sufficient quantities by including in it both heavier and lighter constituents of crude petroleum. They also suggest

50X1-HUM

that kerosene-gas oil fractions obtained by secondary treatment of petroleum and by thermal decomposition or other types of treatment of solid fuels (i.e., products of catalytic cracking, thermal cracking, hydrogenation, the synthin process, etc.) be utilized for the production of diesel fuel. On the basis of the experimental results obtained by them, the authors conclude that the diesel fuels and tractor kerosenes used in the USSR show a somewhat lower performance than the standard fuel used in their investigation. They ascribe the lower performance of the fuels examined to insufficient purification, the presence of unsaturated hydrocarbons, and (in the case of tractor kerosene) addition to the fuel of products of thermal cracking. As one of the most effective remedies improving the quality of diesel fuel they recommend reduction of its acidity by extraction with caustic alkali. They also state that although the use of tractor kerosene was found to result in somewhat rougher operation under certain conditions as compared with the standard fuel, tractor kerosene may still be transformed into a diesel fuel of acceptable quality if the cetane number can be raised to 40-45. In addition to this the authors make several other practical recommendations. They state that on the basis of the work described by them, which involves comparison of acid-free diesel fuel, acid-containing diesel fuel, tractor kerosene, solar oil, and motor oil DT-1 (M_3) with the standard fuel, new standard requirements for diesel fuel as described by GOST (State All-Union Standard) 4749-49 [of attached table] were developed and put into effect.(4)

As far as the acidity of fuel for high speed diesels is concerned, the standard requirements GOST 4749-49 cited by Losikov and Lukashevich do not contain any stipulation in regard to this property (cf. attached table). On the other hand, the USSR handbook of chemical products which was published 4 years later contains an additional requirement in GOST 4749-49 to the effect that the acidity of fuels of this type, as measured in milligrams of potassium hydroxide per 100 milliliters of fuel, must not exceed the value of 5.(8)

[Table showing characteristics of fuels for high-speed diesel engines follows.]

Characteristics of Fuels for High-Speed Diesel Engines (7)

Physical and Chemical Properties	Solar Oil GOST 1666-42	Arctic Diesel Fuel	Winter Diesel Fuel GOST 4749-49	Summer Diesel Fuel GOST 4749-49	Special Diesel Fuel
Cetane Number no Lower Than	--	40	40	45	50
Fractional Composition					
10% distil over at a temperature (in °C) no lower than	--	200	200	--	--
50% distil over at a temperature (in °C) no higher than	--	255	275	290	280
90% distil over at a temperature (in °C) no higher than	--	300	335	350	--
96% distil over at a temperature (in °C) no higher than	--	330	--	--	340
Viscosity at 20°C					
kinematic in centistokes	--	2.5-4.0	3.5-6.0	3.5-8.0	--
E_{20} [degrees Engler?] = conventional degrees	--	1.15-1.30	1.25-1.45	1.25-1.70	--
Viscosity at 50°C					
kinematic in centistokes	2.8-9.0	--	--	--	2.5-4.0
E_{50}	1.2-1.75	--	--	--	1.15-1.3

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[Adjoins page 7 here.]

Coke in %, no More Than	--	0.05	0.05	0.05	0.05
Ash in %, no More Than	0.025	0.01	0.02	0.02	0.02
Sulfur Content in %, no Higher Than	0.2	0.2	0.2	0.2	0.2
Flash Point in °C					
according to Martens-Penskey, no lower than	--	35	50	60	90
according to Brencken, no lower than	125	--	--	--	--
Pour Point in °C, no Higher Than	-20	-60	-45	-10	-15

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